Scanning Tunneling Spectroscopy of $NbSe_2$-Au Proximity Junctions

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(Received 10 March 1999)

The proximity induced excitation spectrum of thin gold layers on superconducting $NbSe_2$ (S) has been measured as a function of gold thickness with a low temperature scanning tunneling microscope. The resulting conductance spectrum is dominated by a quasiparticle bound state below Δ_S with an energy dependence strongly influenced by the suppression of the order parameter in S near the interface. In addition, a significant change in the induced spectrum can be directly related to the sample morphology as determined by imaging. A simple model will be used to interpret the results.

PACS numbers: 74.50.+r, 07.79.Cz

The interface between a normal metal and a superconductor is of both practical and fundamental interest. If the interface is clean and the electronic transmission high, there can be an induced pairing of electrons in the normal metal near the interface. This proximity effect can have a coherence range in the normal metal which can in fact significantly exceed the coherence length in the superconductor [1]. This coherence length enhancement is an important consideration in the fabrication of superconducting devices with short coherence length superconductors, such as superconductor–normal-metal–superconductor (SNS) weak link devices and in superconducting magnet stabilization. While much work has been done on conventional superconductor SN interfaces [2–5], experimental challenges exist which make reliable measurements on superconductors with short coherence lengths, such as $NbSe_2$ and the high-T_c cuprates, difficult to obtain. Such materials have coherence lengths which are not much larger than the Fermi wavelength and exhibit a large anisotropy of the order parameter. These factors challenge the conventional understanding of the proximity effect.

We have found that low temperature scanning tunneling microscopy and spectroscopy, STM/STS, are ideal tools for the investigation of the proximity effect. Direct measurement of the local electronic density of states with high energy resolution can be obtained at different locations on proximity junctions in conjunction with measurements of the sample morphology. The use of an ultrahigh vacuum chamber combined with a low temperature STM and *in situ* sample preparation allows us to fabricate nearly ideal samples. Further sample characterization with the STM gives detailed knowledge of the properties of the metal overlayer. To ensure sample integrity, all measurements are carried out on samples without exposure to the atmosphere.

In this work we present measurements of proximity junctions of thin gold metal layers (N) deposited on a thick single crystal $NbSe_2$ substrate (S). We use electron tunneling to measure the quasiparticle excitation spectrum induced in the N layer, which is related to the N metal thickness and the pair potentials in N and S. The local excitation spectrum is determined by measuring the bias dependent differential conductance of the tunnel junction formed between a Pt STM tip and the gold. Conductance curves are compared for different thickness gold layers and bare $NbSe_2$. The measurements are taken at a temperature of 2.5 K with an energy resolution of better than 100 μV. Great care has been taken to avoid the artificial broadening caused by rf noise which is common to STS measurements.

$NbSe_2$ is a good model superconducting system for study since atomically flat surfaces can easily be prepared by cleaving, and the exposed surface is quite inert. It is an anisotropic superconductor with coherence lengths of 77 Å parallel and 23 Å perpendicular to the layers and a superconducting transition at 7.2 K with a zero temperature superconducting gap of 1.1 meV [6]. Single crystal samples of $NbSe_2$ are cleaved under vacuum and imaged with the STM to determine surface quality. Low temperature measurements of the as-cleaved surface yield atomic resolution images and a BCS excitation spectrum with a gap of 1.0 meV at 2.5 K [7]. Proximity sandwiches are formed by depositing gold layers of average thickness ranging from $d = 0.1$ to 60 nm onto a freshly cleaved $NbSe_2$ crystal held at room temperature. Cooling is achieved by lowering the STM into ultrahigh purity He^4 exchange gas which couples the samples and STM to a liquid helium bath. The temperature is further reduced by pumping the bath to reach 2.5 K. Differential conductance measurements are made using the ac lock-in technique with a 50–100 μV ac component added to the dc sample bias. Conductance measurements are compared for different tunneling resistances over the range of 10^8 to 10^9 Ω, and no dependence on tunneling resistance is evident. The current was also measured as a function of tip height and the resulting $I(z)$ curves were exponential, indicating true vacuum tunneling.

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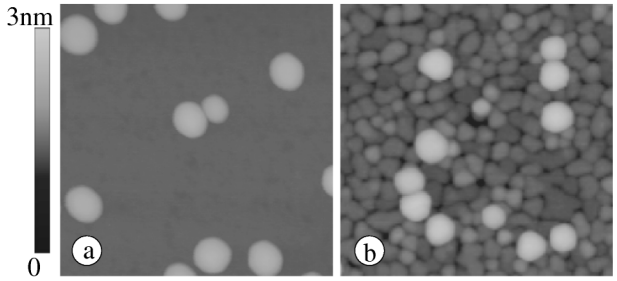

FIG. 1. (a) 100 nm image of 0.1 nm gold on $NbSe_2$; (b) same sample after incremental growth to 4 nm gold.

Isolated grains have been obtained by depositing gold with an average film thickness less than a monolayer ($d = 1$ nm) onto the $NbSe_2$ surface. The gold clusters into islands of about 2.5 nm thickness and 10 nm diameter separated by large areas of bare $NbSe_2$ (Fig. 1a). Tunneling into the surface far from any gold reveals an excitation spectrum identical to that obtained on the bulk $NbSe_2$ sample (lowest curve in Fig. 2), having the normal BCS shape and an energy gap of 1.0 meV at 2.5 K. Tunneling directly into one of the gold grains of Fig. 1a produces a conductance curve with a proximity induced gap of 1.0 meV, which is of the same magnitude as observed on the bare superconductor, indicating that the grain is fully proximity coupled to the superconductor (Fig. 2, 0.1 nm). As more gold is deposited onto the surface shown in Fig. 1a, the density of grains increases until the point where grain stacking appears (Fig. 1b).

Tunneling results for each average thickness are shown in Fig. 2. We observe a decrease in the magnitude of the gap as the thickness of the metal layer is increased, as one would expect. For a 10 nm film, the gap has been reduced to $\approx$0.6 meV. It should be emphasized that the morphology of this film is that of stacked pancake shaped particles of about 2.5 nm thickness, so that the 10 nm film is roughly four particles thick. Conductance curves measured on the gold exhibit a conductance peak near the gap edge which is somewhat sharper than that predicted

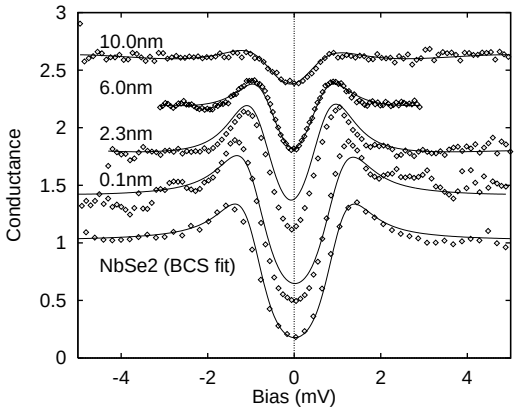

FIG. 2. Normalized conductance measured at $T = 2.5$ K for the samples of Fig. 1 at different gold thicknesses (as indicated). Solid lines are proximity model best fits.

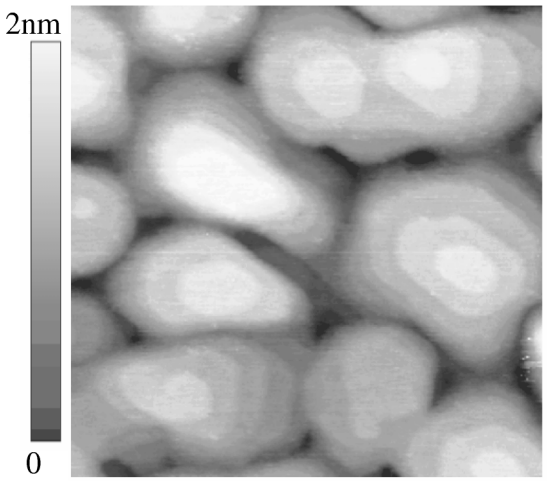

FIG. 3. 100 nm image of a 4-nm-thick gold film deposited as an initial 1 nm film followed by successive increments.

by a simple BCS model. This sharp conductance peak is due to quasiparticle interference effects resulting in a bound state near the gap edge [8,9].

A striking change in the morphology is apparent for samples which begin with an initial gold thickness great enough to completely cover the $NbSe_2$ ($d = 1$ nm). These samples exhibit much larger structures, with typical lateral dimensions of 40–50 nm and atomic steps clearly visible at the surface (Fig. 3). The deposition rate is unchanged at 0.2 Å/s from the results shown in Fig. 1. The only difference between the growth conditions is that the first 1 nm or more is deposited in one step with no interruption. Tunneling results also show a marked change over the samples of Fig. 1. Figure 4 shows conductance curves for gold thicknesses up to $d = 60$ nm. A significantly different dependence on the metal thickness is found. An energy gap is measurable for tunneling into gold of thickness greater than 60 nm as compared to about 10 nm as shown in Fig. 2. Gap edges sharper than BCS are also observed for all thicknesses (see the discussion below).

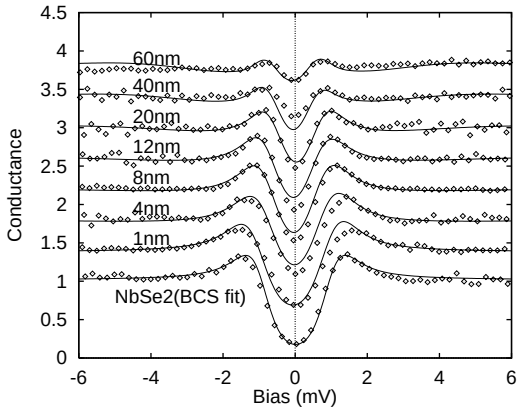

FIG. 4. Normalized conductance measured at $T = 2.5$ K for the samples of Fig. 3 at different gold thicknesses (as indicated). Solid lines are proximity model best fits.

One may be tempted to reason that the observed energy gap is due to a finite pair potential induced in the normal metal, Δ_N, resulting in a BCS-like excitation spectrum at the surface of N. For sufficiently thin N and S layers ($d_N \ll \xi_N, d_S \ll \xi_S$), where the spatial variation of the order parameter can be neglected, this approximation gives reasonable results (Cooper limit) [10]. However, the more general case of thicker N and S layers is somewhat more involved since the order parameter can have a significant spatial dependence. This variation is particularly important when considering superconductors with short coherence lengths ($\xi_S \approx 10$ Å), since the corresponding Cooper limit is experimentally difficult to reach. The case of a semi-infinite S layer with a thin N layer can be more accurately described by considering the pair potential variation across the NS interface and calculating the energy of quasiparticle states in this potential, the de Gennes-St. James bound states [8]. One may consider the simplest pair potential variation as a potential well of depth $\Delta_S - \Delta_N$ and width d. One can show that when the size of this well is small, $d \ll \frac{\hbar v_{FN}}{2\Delta_S}$, only one bound state level E_0 exists, and its energy approaches Δ_S as d is reduced to zero [8]. A consequence of this bound quasiparticle state is that, even though there may be a finite pair potential in N, there may not be any BCS-like peak at Δ_N due to the quantization imposed by confinement to a finite potential well [11].

It is worth considering the difference between a BCS spectrum and that derived from a bound state level model. Figure 5 shows a comparison of data for two gold thicknesses (8 and 60 nm) and "best fits" using a simple BCS model with a variable gap energy and the conductance spectrum of a bound state level at $E_0 < \Delta_S$ with $\Delta_S = 1.0$ meV. The bound state model produces sharper conductance peaks and a dip in conductance at energies above the peak for thicker N metal films. From this comparison we are drawn toward the conclusion that we are observing a proximity spectrum dominated by the bound state.

We have chosen to fit the data with the Arnold proximity tunneling model which predicts a quasiparticle excitation spectrum based on the pair potentials in N and S and the energy of the resonant bound state, E_0 [9]. We make the assumption that $\Delta_N = 0$, which is reasonable for weak coupling materials such as gold [9], and let $\Delta_S = 1.0$ meV. The fit is then optimized by adjusting the value of E_0 to minimize the least square error. The best fit conductance curves are plotted as solid lines in Figs. 2 and 4. The resulting curves represent the conductance of a quasiparticle bound state with an energy $E_0 < \Delta_S$ in a well of depth Δ_S. The assumption that Δ_S is constant is crude as it implies an abrupt change in the pair potential in S near the interface, which generally is not the case. A detailed calculation of the bound state levels of an arbitrary spatially varying pair potential is rather difficult; however, one can reach some understanding from

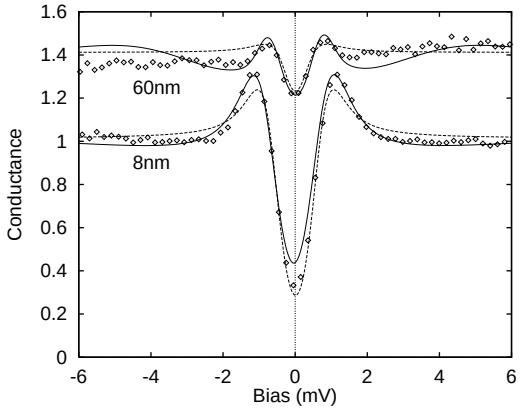

FIG. 5. Comparison of proximity bound state results (solid line) with a simple BCS spectrum (dashed line) for two gold thicknesses.

the simplified case of a rectangular well. For a well of depth Δ_S and width d in the limit of small d, the first bound state energy is given by $E_0 \approx \Delta_S[1 - \frac{1}{2}(\frac{2\Delta_S d}{\hbar v_F})^2]$. Since $d \ll \hbar v_F/2\Delta_S = 440$ nm in the present measurements, it is clear that the bound state energy observed in these measurements is significantly lower than expected from a state confined to the normal metal layer only (well of width d_N) by observing that E_0 drops well below Δ_S for N metal thicknesses less than 60 nm as shown in Fig. 6. This implies that the quasiparticles spend a significant fraction of their time in the pair potential suppressed region of the superconductor ($NbSe_2$), as has been previously suggested by Tessmer *et al.* [12,13]. This pair potential suppressed region extends a distance of the order of $\xi_S = 23$ Å into the superconductor, which is comparable to the metal thickness. However, because the c-axis Fermi velocity for $NbSe_2$ is much less than gold [14], $v_{FS}/v_{FN} \ll 1$, the quasiparticles may spend a significantly longer time in the potential suppressed region of S than in N, even when the metal thickness exceeds ξ_S. The result is a larger effective confinement length for the quasiparticles producing a lower bound state energy level. As the thickness of the normal metal layer is increased,

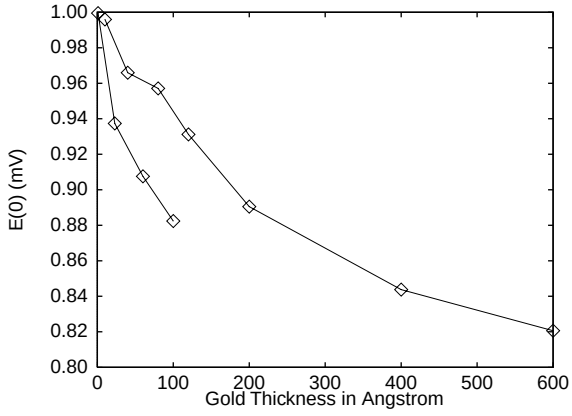

FIG. 6. Best fit bound state energy versus gold thickness.

the pair potential in S is more strongly suppressed near the interface, increasing the length over which quasiparticles with energy below Δ_S may extend into the superconductor, further reducing the bound state energy. While previous measurements suggested that the order parameter is almost completely suppressed in $NbSe_2$ by a thin (4 nm) layer of gold [12,13], our results indicate that the suppressed order parameter in S is finite for a gold layer thickness up to at least 60 nm. The difference between these two experimental results can partially be understood as due to the difference in the gold film morphology. The isolated grains of Fig. 1 are smaller in size and more regular than those reported previously. These smaller grains would produce a smaller suppression of the order parameter in the superconductor. The thicker and more uniform metal films of Fig. 3 further emphasize the strong dependence of the tunneling characteristic on the film structure. Furthermore, no fine structure appears in our STM images. This implies both smooth regular grains and well-defined tunneling tips for our STM.

The difference between the data sets shown in Figs. 2 and 4 likely arises from the higher grain boundary density of the samples shown in Figs. 1 and 2 which leads to a shorter scattering time, a shorter equivalent mean free path, and, hence, a shorter coherence length than for a continuous film. This will broaden the energy of the bound state and smear the conductance spectrum. Increased elastic scattering can also lead to a greater effective quasiparticle confinement length, reducing the bound state energies [11]. This can most easily be seen by considering the average distance an electron must traverse to pass from the superconductor through the metal layer to the STM tip. If the film is highly granular, as shown in Fig. 1b, scattering at the grain boundaries of grains stacked in the perpendicular direction increases the time an electron spends in each grain before traversing to another and therefore the total effective path length the electron must traverse through the metal film. It is clear from these results that the structure of the metal layer can greatly affect the nature of the proximity effect.

While the analysis shown here is clearly oversimplified, we believe it indicates that the short coherence length in $NbSe_2$ and a low Fermi velocity are important considerations for determination of the proximity induced excitation spectrum. Current proximity effect models make assumptions about both S and N materials which may not be reasonable when one of the materials has a very short coherence length, such as in the case of $NbSe_2$ and high-T_c superconductors. Another important factor is that $NbSe_2$ has a Fermi surface quite different from that of a conventional metal [14], with a reduced Fermi velocity in the direction normal to the surface. The Fermi mismatch at the interface introduces an enhanced normal reflection which would tend to suppress the proximity effect [11]. These properties require a more sophisticated model than has been applied here to fully understand the nature of the proximity effect between $NbSe_2$ and gold and similarly with high-T_c materials such as $Bi_2Sr_2CaCu_2O$.

In summary, we have successfully measured the proximity induced excitation spectrum for a number of Au-$NbSe_2$ proximity junctions. The results indicate an energy spectrum which is dominated by a bound state level with an energy which strongly depends on the suppression of the order parameter in $NbSe_2$. In addition, this proximity suppression has been shown to depend strongly on the structure of the normal metal film, with a stronger effect being observed for more granular film. This technique is applicable to some of the cuprate superconductors and results from these measurements will be forthcoming [15].

The authors wish to thank G. B. Arnold, D. Arovas, H. F. Hess, and A. Frydman for helpful discussion. This work is supported by the Office of Naval Research Grant No. N00014-97-1-0897.

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